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**PROPERTIES OF UNATTACHED  
POLONIUM-218 IN THE  
PROPYLENE-NITROGEN GAS SYSTEM**

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ABSTRACT

Electrostatic and diffusion properties of unattached polonium 218 were investigated in a propylene-nitrogen gas system. The diffusion coefficient of neutral unattached polonium 218 under various relative humidities was measured and was found to be lowered by humidity owing to the formation of clusters. The fraction of polonium 218 having a positive charge at a gas age of about 0.1 second was determined. The mobility of the charged polonium 218 was obtained independently by using a pulse width modulated ion mobility analyzer, which employed the time of flight method. The diffusion coefficient and mobility data obtained fitted the Einstein equation on the assumption that the polonium 218 ion has one positive charge. The neutralization rate constant of polonium 218 ions under various relative humidities and radon concentrations was investigated, and three neutralization mechanisms were observed. The neutralization rate constant is proportional to the 0.2 power of the radon concentration at all relative humidities, suggesting that the small ion recombination mechanism plays an important role. The charge transfer mechanism involving propylene and polonium hydroxide ions is responsible for lowering the power dependence of the neutralization rate constant on radon concentration from 0.5 (in nitrogen) to 0.2 (in propylene-nitrogen). The presence of the electron scavenging mechanism involving hydroxyl groups formed from water vapour radiolysis causes the neutralization rate constant to increase with relative humidity.

RÉSUMÉ

Le présent rapport examine les propriétés électrostatiques et diffusionnelles du polonium 218 libre dans un mélange gazeux de propylène et d'azote. On a mesuré le coefficient de diffusion du polonium 218 libre neutre pour différentes valeurs de l'humidité relative et on a trouvé qu'il décroît lorsque l'humidité relative augmente, du fait de la formation d'agglomérats. On a déterminé la fraction de polonium 218 chargée positivement dans un mélange gazeux âgé de 0,1 seconde. La mobilité électrique du polonium 218 chargé a été déterminée indépendamment, grâce à un analyseur de mobilité ionique à impulsions de durée variable, en employant la méthode du temps de vol. Les résultats obtenus concernant les coefficients de diffusion et la mobilité sont en accord avec l'équation d'Einstein si l'on suppose que l'ion de polonium 218 ne porte qu'une seule charge élémentaire. Le taux de neutralisation du polonium 218 chargé a été étudié sous différentes conditions d'humidité relative et de concentration du radon, et on a observé trois mécanismes de neutralisation différents. Le taux de neutralisation est proportionnel à la puissance 0,2 de la concentration du radon pour toute valeur de l'humidité relative. Ceci suggère que la recombinaison des petits ions joue un rôle important. Dans un mélange de propylène et d'azote, le taux de neutralisation devient moins dépendant de la fonction de puissance de la concentration du radon, l'exposant passant de 0,5 pour l'azote à 0,2 pour le mélange de propylène et d'azote, en raison d'un mécanisme de transfert de charge mettant en cause la molécule de propylène et l'ion d'hydroxide de polonium. Des groupes hydroxyles formés par radiolyse de la vapeur d'eau créent des mécanismes avides d'électrons, d'où un accroissement du taux de neutralisation avec l'humidité relative.

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## Introduction

The first progeny in the radon decay chain is polonium-218, which is formed from the  $\alpha$ -decay of radon-222. Wellisch<sup>(1)</sup> found that 88.2% of polonium-218 formed singly charged positive ions in a dry air system. The process of neutralization of polonium-218 occurs after formation of the ion, and affects deposition and transport of the progeny in various environments.

Various methods have been used to measure the diffusion coefficient of polonium-218 in air and in gas mixtures containing trace gases such as water vapour or nitrogen dioxide. Wellisch<sup>(1)</sup> found that the neutral polonium-218 species had a diffusion coefficient of 0.045 in dry air. Raabe<sup>(2)</sup> used a diffusion tube technique to determine the diffusion coefficient of polonium-218 at various relative humidities (R.H.). Neutral polonium-218 was found to have a diffusion coefficient of 0.047 cm<sup>2</sup>/s at R.H. = 15% and 0.034 at R.H. = 35%. Thus the diffusion coefficient was found to decrease with R.H. Thomas and LeClare<sup>(3)</sup> used the two-filter method and found that the diffusion coefficient increased from 0.053 cm<sup>2</sup> at R.H. = 0% to 0.085 cm<sup>2</sup>/s at R.H. = 20%. The difference in the results is probably due to the difference between the ages of the polonium species in the experiments of Raabe and Thomas. The age of Raabe's polonium-218 was of the order of a few minutes, whereas Thomas' polonium-218 had an average age of 15-29 sec. The lower diffusion coefficients found by Raabe at higher humidities are probably the result of the growth of clusters.

The diffusion coefficient of a charged atom is smaller than that of a neutral atom. Porstendorfer and Mercer<sup>(4)</sup> reported that the diffusion coefficient of charged polonium 218 in air was 0.024 cm<sup>2</sup>/s and that of the neutral species 0.068 cm<sup>2</sup>/s.

Busigin et al.<sup>(5)</sup> used an electrostatic collection apparatus and found the diffusion coefficient of neutral polonium-218 species in argon.

At R.H. = 8%,  $D_A = 0.028 \text{ cm}^2/\text{s}$ ; at R.H. = 15%,  $D_A = 0.043 \text{ cm}^2/\text{s}$ ; and at R.H. = 100%,  $D_A = 0.046 \text{ cm}^2/\text{s}$ .

Goldstein and Hopke<sup>(6)</sup> determined the diffusion coefficient of unattached polonium-218 in dry oxygen and in nitrogen in the presence of the trace gases  $n\text{-C}_4\text{H}_{10}$ ,  $\text{C}_5\text{H}_{12}$ ,  $\text{NH}_3$ ,  $\text{NO}_2$  and  $\text{NO}$ . The diffusion coefficient for neutral polonium-218 was determined to be  $0.072 \text{ cm}^2/\text{s}$ , and for a mixture of charged and neutral species,  $0.037 \text{ cm}^2/\text{s}$ .

Thus, based on several investigations, the diffusion coefficient of polonium-218 has a particular value for a given set of conditions, especially the age and charged state of the species, the relative humidity, and the nature and concentration of any trace gases.

Little work has been carried out on the neutralization rate of polonium-218. Busigin et al.<sup>(5)</sup> noted that polonium ions are chemically unstable and may react with oxygen in the atmosphere to form oxide ions,  $\text{PoO}_2^+$ , and estimated the ionization potential of  $\text{PoO}_2^+$  to be about 10 eV. Subsequently, Goldstein and Hopke<sup>(6)</sup> estimated the ionization potential of  $\text{PoO}_2^+$  to be 10.3-10.53 eV.  $\text{PoO}_2^+$  ions may be neutralized by transferring charge to trace gases such as nitrogen dioxide and propylene, which have lower ionization potentials<sup>(5)</sup>.

Busigin et al.<sup>(5)</sup> found small ion recombination to be an important neutralization mechanism for polonium-218 ions. By assuming that neutralization of polonium-218 ions occurs primarily through recombination with negative small ions, Busigin et al.<sup>(5)</sup> predicted that the recombination rate should be proportional to the square root of the radon concentration. Later,

Chu and Hopke<sup>(7)</sup>, using a pure nitrogen system, verified that the neutralization rate of small ion recombination varied as the square root of the radon concentration, as predicted by Busigin et al.

Leung and Phillips<sup>(8)</sup> concluded that the neutralization rate constant in a pure argon atmosphere is relatively independent of radon concentration at low radon concentrations ( $<10^5$  atoms/cm<sup>3</sup>), but proportional to the 0.6 power of radon concentration at high radon concentrations ( $>10^5$  atoms/cm<sup>3</sup>). In an argon-nitrogen dioxide gas system, Leung and Phillips found that at radon concentrations less than  $10^5$  atoms/cm<sup>3</sup>, the neutralization rate constant is independent of radon concentration, and at radon concentrations greater than  $10^5$  atoms/cm<sup>3</sup>, the rate constant is proportional to the 0.3 power of radon concentration. Leung and Phillips<sup>(8)</sup> suggested that the dominant neutralization process at low radon concentration ( $<10^5$  atoms/cm<sup>3</sup>) is charge transfer and that, at high radon concentration ( $>10^5$  atoms/cm<sup>3</sup>), it is ion recombination.

The objective of the present study was to determine the fundamental electrostatic and diffusion properties of unattached polonium-218 in the presence of trace propylene gas, and to investigate the mechanisms of neutralization under different conditions. Important conditions include those involving hydrocarbon gases at low concentrations, since hydrocarbons are commonly present in polluted air. Propylene was selected as the hydrocarbon gas to be studied because of its relatively low ionization potential of 9.73 eV<sup>(9)</sup>.

### Experimental

An electrostatic collection apparatus and a pulse width modulated ion

mobility analyzer were used to perform the experiments. The electrostatic collection apparatus consists of two parallel aluminum discs separated by a Teflon ring<sup>(8)</sup>. The potential of the top plate is held at ground and that of the bottom plate is varied (positive or negative) to allow collection of positively charged species on either the top or bottom plate. Neutral species are collected by diffusion. A ruggedized silicon surface barrier detector (E.G.&G. Ortec R series) is located in the centre of the top plate to monitor the process without disturbing the uniform electric field.

Radon gas was produced by emanation from radium chloride solution in a bubbler. The relative humidity was measured by a YSI dewpoint hygrometer. The pressure inside the apparatus was atmospheric, and was checked by measurement for each experiment.

In order to obtain independent mobility data for the charged polonium-218 species, the pulse width modulated ion mobility analyzer, employing the time of flight method<sup>(8)</sup>, was connected to the electrostatic collection apparatus.

The flux of polonium-218 species to the detector surface was measured by an alpha spectrometer system (Canberra). The 6.0 MeV  $\alpha$  energy of polonium-218 was separated from the 7.69 MeV  $\alpha$  energy of polonium-214 and the 5.48 MeV  $\alpha$  energy of Rn-222.

The electrostatic collection model developed by Leung and Phillips<sup>(8)</sup> was employed to evaluate the four parameters,  $D_A$ ,  $F$ ,  $C_{Rn}$  and  $K/B$ . The model is based on differential equations describing steady state transport of charged polonium-218 in the presence of electric field, and steady state diffusive transport of both charged and neutral polonium-218 species in the absence of an electric field. Flux equations are obtained at different

electric field strengths. The count rate, which is proportional to the flux of polonium-218 at steady state, is fitted as a function of the applied electric field to yield the four parameters  $D_A$ ,  $F$ ,  $C_{Rn}$  and  $K/B$ . An optimization procedure<sup>(10)</sup> based on an iterative, weighted least squares fitting routine in four dimensions is used for parameter evaluation. The equations involved have been described elsewhere<sup>(8)</sup>.

Experiments were carried out to determine the electrostatic and diffusive properties of unattached polonium-218 in the presence of trace amounts of propylene in a nitrogen gas system. The experimental work consisted of two phases. Firstly, the electrostatic collection apparatus was used to determine the diffusion coefficient  $D_A$ , the fraction born charged  $F$ , and the ratio of the neutralization rate constant  $K$  to the mobility,  $B$  (that is  $K/B$ ) of unattached polonium-218 in the propylene/nitrogen system. Secondly, a pulse width modulated mobility analyzer was used to obtain independent mobility data, from which the neutralization constant was calculated.

The results for  $D_A$ ,  $F$ ,  $B$  and  $K$  for polonium-218 ions in a propylene-nitrogen gas system are shown in Table 1.

#### Diffusion Coefficient of Neutral Unattached Po-218, $D_A$

The overall average  $D_A$  is  $0.041 \text{ cm}^2/\text{s}$  (relative standard deviation of about 25%). The results for  $D_A$  are comparable to values reported by Goldstein and Hopke<sup>(6)</sup> for alkanes, namely  $0.0355$  in  $1.5 \text{ ppm C}_5\text{H}_{10}$  in  $\text{N}_2$  and  $0.0365$  in  $16 \text{ ppb n-C}_5\text{H}_{12}$  in  $\text{N}_2$ .

In gas systems of R.H. = 96-100%, the average diffusion coefficient was found to be  $0.023 \text{ cm}^2/\text{s}$ . A lower value is expected because of the formation of clusters.

The diffusion coefficient was found not to vary with the concentration of propylene over the range 10-1000 ppm, showing that complete neutralization of neutral unattached Po-218 occurs at concentrations less than 10 ppm of propylene in nitrogen.

#### Fraction Born Charged, F

The fraction of Po-218 having a positive charge at the end of the recoil path, F, was found to have an average value of 0.62, in agreement with the fraction born charged value reported by Leung and Phillips<sup>(8)</sup> in an argon gas system at a gas age of 0.1 s. The values of F were found to be constant (within  $\pm 6\%$ ) and independent of radon concentration in the propylene-nitrogen system.

The positive fraction was found to decrease as the relative humidity increased (Fig. 1). Since the collection time in the experiments was of the order of 0.1 s, neutralization would occur between the end of recoil and the end of the 0.1 s interval. The neutralization rate was found to increase with relative humidity.

The average value of F (0.62) was smaller than that reported by Porstendorfer and Mercer<sup>(4)</sup> (0.85-0.88) because the collection time in the present experiments was about 0.1 s, whereas the collection time in the work of Porstendorfer and Mercer was of the order of a few msec. The value of F found in the present system suggests that various neutralization reactions may occur between the end of the recoil path and the end of the collection time interval. The age of the gas system would thus be important in determining the fraction born charged. The average value found, namely 0.62, reflects only the unattached charged fraction of Po-218 at a gas age of about

0.1 s.

#### Mobility of the Charge Unattached PO-218. B

In the present work, the time of flight method was employed to determine independent mobilities. The neutralization rate was calculated for each experiment. Mobilities of the PO-218 ions were determined to be between 1.08 cm<sup>2</sup>/s.V in a humid gas system (R.H. = 100%) and 1.80 cm<sup>2</sup>/s.V in a dry gas system (R.H. = 15%). The average value found for B was 1.51 cm<sup>2</sup>/s.V, which is comparable to values reported by Chu and Hopke<sup>(7)</sup> (gas system: nitrogen; 1.87 cm<sup>2</sup>/s.V), Leung and Phillips<sup>(8)</sup> (gas system: argon; 0.7 to 1.4 cm<sup>2</sup>/s.V), and Busigin et al.<sup>(5)</sup> (4) (gas system: nitrogen; 2.04 cm<sup>2</sup>/s.V).

The difference between the average experimentally determined mobility (1.51 cm<sup>2</sup>/s.V) and the theoretical mobility for freshly formed Po-218 ion (2.04 cm<sup>2</sup>/s.V, as calculated from kinetic theory<sup>(5)</sup>), suggests that clustering and charge exchange interactions could occur during the collecting time interval, which is of the order of 0.1 s. Figure 2 shows that the mobility of Po-218 ion decreases with increasing relative humidity, consistent with the occurrence of clustering, as previously reported by Leung and Phillips<sup>(8)</sup>.

The mobility can also be calculated from the Einstein equation<sup>(11)</sup>:

$$\frac{B}{D} = \frac{e}{kT}$$

where B = mobility

D = diffusion coefficient

e = elementary charge ( $= 1.60 \times 10^{-19}$  coulomb)

k = Boltzmann's constant ( $= 1.38 \times 10^{-23}$  J/K)

T = absolute temperature.

Based on the average diffusion found in the present study (0.041 cm<sup>2</sup>/s),

the mobility estimated from the Einstein equation is  $1.61 \text{ cm}^2/\text{s.V}$ , on the assumption that the polonium-218 ion has one positive charge. The deviation between the calculated value (1.61) and the average experimental result (1.51) is only 6%, which is probably not significant. An alternative interpretation based on the good agreement between the calculated and the experimental is that polonium-218 ions are singly charged. This result is in agreement with reports in the literature<sup>(4)</sup>.

#### Neutralization Rate Constant, K, for Charged Unattached Po-218 ions

In the present work, radon concentration, relative humidity and trace gas (propylene) concentration were varied to investigate their effect on the neutralization rate constant of polonium-218 ions.

The neutralization rate constant was measured in radon concentrations from  $1.5 \times 10^4$  to  $5.9 \times 10^6 \text{ atom/cm}^3$  in 10 ppm propylene (balance nitrogen) at various relative humidities. At R.H. = 12-17%, K increased from 16 to 60  $\text{sec}^{-1}$ ; at R.H. = 38-45%, K increased from 22 to 75  $\text{sec}^{-1}$ , and at R.H. = 96-100%, K increased from 31 to 85  $\text{sec}^{-1}$ . A plot of K vs  $C_{\text{Rn}}$  at various relative humidities (Fig. 3) shows that the neutralization rate constant of polonium-218 ions increases with both radon concentration and relative humidity.

Chu and Hopke<sup>(7)</sup> reported that in pure nitrogen the small ion recombination rate of polonium ions followed a linear relation:

$$K = 0.0176 C_{\text{Rn}}^{0.5} - 1.613$$

This equation implies that K is negative at low radon concentrations, which is not possible. The likely explanation is that diffusion and plate-out are significant and dominate in the mixture investigated, so that a minimum radon

concentration is required before small ion recombination can be observed. Nolan<sup>(12)</sup> has previously pointed out the importance of diffusion and plate-out in small systems.

In the present work, the neutralization rate constant was correlated with radon concentration,  $C_{Rn}$ , as follows<sup>(7)</sup>:

$$K = 4.25 C_{Rn}^{0.19} - 2.38 \quad \text{at R.H.} = 96-100\%$$

$$K = 2.72 C_{Rn}^{0.21} - 1.89 \quad \text{at R.H.} = 38-45\%$$

$$K = 1.76 C_{Rn}^{0.23} - 2.11 \quad \text{at R.H.} = 12-17\%$$

Correlation coefficients for linear regression were 0.996, 0.988 and 0.992 respectively. As noted above, the constant in these equations suggests that plate-out is significant, and that a minimum radon concentration is required in order that neutralization can become effective. Thus the neutralization rate constant can be considered to be proportional to the one fifth power of the radon concentration. Busigin et al.<sup>(5)</sup> predicted that the recombination rate should be proportional to the square root of radon concentration. If the small ion recombination mechanism dominates the neutralization process, the neutralization rate constant will be proportional to the negative small ion concentration,  $C_-$ . The negative small ion concentration depends on gas ionization reactions according to the following equation:

$$\frac{dC_-}{dt} = \xi C_{Rn} - \gamma C_+ C_-$$

where  $\gamma$  is the recombination coefficient of negative and positive small ions

$\xi$  is the rate constant for small ion production

$C_+$  is the positive small ion concentration.

If it is assumed that  $C_+ = C_-$ , then, at steady state, as shown by Busigin et al.<sup>(5)</sup>:

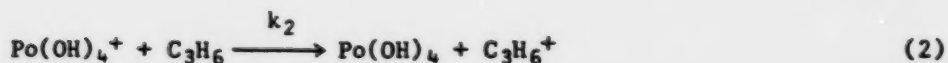
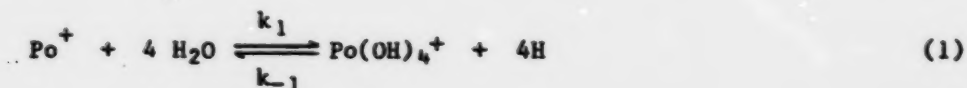
$$C_- = \sqrt{\frac{\xi C_{\text{Rn}}}{\gamma}}$$

Thus the negative small ion concentration  $C_-$ , and hence the neutralization rate constant,  $K$ , is proportional to the square root of the radon concentration:

$$k = A C_{\text{Rn}}^{0.5}$$

In the present experiments, charge transfer co-exists with small ion recombination. Since the charge transfer rate constant would decrease with increasing radon concentration, this negative effect would lower the dependence of the neutralization rate constant on the power of the radon concentration. The 0.2 power exponent found in the present work suggests that, in addition to ion recombination, charge transfer also occurs in the neutralization of polonium-218. In other words, charge transfer causes the difference in the exponent between the results of Busigin et al.<sup>(5)</sup> (0.5) and the present study (0.2).

The presence of propylene with an ionization potential of 9.73 eV would probably cause the following charge transfer reaction in which the  $k_i$  are the rate constants:



The charge transfer neutralization rate constant may be calculated as follows, using the convention of square brackets to indicate concentration:

$$\begin{aligned} \frac{d[\text{Po}(\text{OH})_4^+]}{dt} &= k_1[\text{Po}^+][\text{H}_2\text{O}]^4 - k_{-1}[\text{Po}(\text{OH})_4^+][\text{H}]^4 \\ &\quad - k_2[\text{Po}(\text{OH})_4^+][\text{C}_3\text{H}_6] \end{aligned}$$

By assuming steady state for  $\text{Po}(\text{OH})_4^+$ , it follows that

$$[\text{Po}(\text{OH})_4^+] = \frac{k_1[\text{Po}^+][\text{H}_2\text{O}]^4}{k_{-1}[\text{H}]^4 + k_2[\text{C}_3\text{H}_6]}$$

According to process (2),

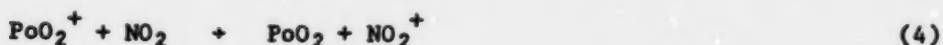
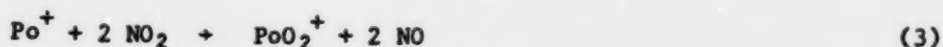
$$\begin{aligned} \frac{d[\text{Po}(\text{OH})_4]}{dt} &= k_2[\text{Po}(\text{OH})_4^+][\text{C}_3\text{H}_6] \\ &= \frac{k_2 k_1[\text{Po}^+][\text{H}_2\text{O}]^4[\text{C}_3\text{H}_6]}{k_{-1}[\text{H}]^4 + k_2[\text{C}_3\text{H}_6]} \end{aligned}$$

Hence the neutralization rate constant is given by

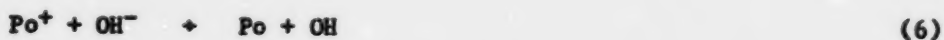
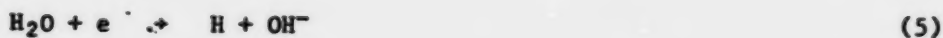
$$K = \frac{k_1 k_2[\text{H}_2\text{O}]^4[\text{C}_3\text{H}_6]}{k_{-1}[\text{H}]^4 + k_2[\text{C}_3\text{H}_6]}$$

Thus, the neutralization rate constant from charge transfer should increase with water vapour concentration and decrease with hydrogen concentration. The hydrogen atom is mainly produced by the radiolytic decomposition of water vapour. When the radon concentration increases, the radiolytic rate of water vapour decomposition increases,  $[\text{H}_2\text{O}]$  decreases and the  $[\text{H}]$  increases. As a result, an increase in radon concentration results in a decrease in the neutralization rate of the charge transfer process. The difference in the exponent between the work of Busigin et al.<sup>(5)</sup> (4) (0.5) and the present study (0.2) may be explained on this basis. As the radon concentration increases, the negative effect from charge transfer neutralization would lower the rate of increase of the neutralization rate constant.

The 0.2 power dependence results from both small ion recombination and charge transfer. Leung and Phillips<sup>(8)</sup> reported a 0.3 power dependence in the nitrogen dioxide-argon gas system. Since the ionization potentials of propylene (9.73 eV) and nitrogen dioxide (9.79 eV) are virtually the same, the exponent difference between the propylene-nitrogen system (0.2) and the nitrogen dioxide-argon (0.3) system is probably because propylene does not react with polonium-218, whereas nitrogen oxide reacts with polonium-218 ions according to:



The polonium dioxide ions (ionization potential ~10.5 eV) formed initially undergo charge transfer with nitrogen dioxide (ionization potential = 9.79 eV). This neutralization process would increase the power dependence of K on  $C_{\text{Rn}}$ . The neutralization rate constant was found to increase with relative humidity, in agreement with reports of Leung and Phillips<sup>(8)</sup> and Goldstein and Hlopke<sup>(6)</sup>. The electron scavenging process involving water vapour may be expressed as follows:



The low electron affinity of the hydroxyl radical (1.83 eV) causes the OH radical produced from water vapour radiolysis to be an excellent electron acceptor and a significant reactant in neutralizing polonium ions. The propylene concentration seemed to have no significant effect on the neutralization rate constant, suggesting that the lowest propylene concentration in this study, 10 ppm, was more than sufficient for the completion of the charge

transfer process involving propylene, namely, processes 1 and 2.

In the present study, the relationships between the neutralization rate constant and the relative humidity, and between the neutralization rate constant and the radon concentration were similar to those found by Leung and Phillips<sup>(8)</sup> and by Chu and Hopke<sup>(7)</sup>. Three neutralization mechanisms may be inferred:

(1) small ion recombination: the charged polonium-218 ions are neutralized by recombination with small negative ions (including electrons).

(2) charge transfer: the charged polonium-218 reacts with water vapour to form polonium hydroxide ions, which are then neutralized by propylene through charge transfer to form polonium hydroxide and the propylene ion (reactions (1) and (2)).

(3) electron scavenging via hydroxyl groups: the hydroxyl radical from water vapour radiolysis scavenges electrons to form the hydroxide ion, which then neutralizes the polonium ions.

### Conclusions

In the propylene-nitrogen gas system, the diffusion coefficient has an average value of  $0.041 \text{ cm}^2/\text{s}$ , comparable to values reported by Goldstein and Hopke<sup>(6)</sup> for alkane systems.

The mobilities of polonium-218 ions were determined to be  $1.08 \text{ cm}^2/\text{s.V}$  at R.H.  $\sim 100\%$  and  $1.80 \text{ cm}^2/\text{s.V}$  at R.H.  $\sim 15\%$ . The average mobility found,  $1.51 \text{ cm}^2/\text{s.V}$ , is in agreement with values in the literature (4,6,7). Based on the assumption of a single positive charge on the polonium-218 ions, the mobility estimated from the Einstein equation is  $1.61 \text{ cm}^2/\text{s.V}$ , which is within 6% of the average experimental value of  $1.51 \text{ cm}^2/\text{s.V}$ . Based on the

close agreement between the calculated and experimental values, the data may alternatively be interpreted as indicating that the polonium-218 ions are singly charged. The fraction of polonium-218 having a positive charge at a gas age of 0.1 sec was found to be 62%, in agreement with the value found by Leung and Phillips<sup>(8)</sup> for an argon gas system.

The neutralization rate constant was found to be proportional to the 0.2 power of the radon concentration, suggesting that small ion recombination plays an important role in the neutralization of the unattached polonium-218 positive ions.

The charge transfer mechanism involving propylene and polonium hydroxide ions lowers the power dependence of the neutralization constant on radon concentration from 0.5 (in nitrogen) to 0.2 (in propylene-nitrogen). Since the charge transfer neutralization rate increases with  $[H_2O]$  and decreases with  $[H]$ , an increase in radon concentration causes a decrease in the charge transfer neutralization rate due to the increasing rate of radiolytic decomposition of water vapour.

The neutralization rate constant was found to increase with relative humidity, suggesting that the electron scavenging process involving water vapour radiolysis is an important neutralization mechanism.

The neutralization of unattached polonium-218 positive ions is a complicated process, and is controlled by different mechanisms under different conditions. The three neutralization mechanisms observed (small ion recombination, charge transfer and electron scavenging) all play important roles in neutralizing charged polonium-218 ions.

## References

1. Wellisch, E.M. (1913). The Distribution of the Active Deposit of Radium in an Electric Field - II. *Philos. Mag.*, 26, 623.
2. Raabe, O. (1968). Measurement of the Diffusion Coefficient of RaA. *Nature (London)*, 217, 1143.
3. Thomas, J.W. and LeClaire, P.C. (1970). A Study of the 2-filter Method for Radon-222. *Health Phys.*, 18, 113.
4. Porstendorfer, J. and Mercer, T.T. (1979). Influence of Electric Charge and Humidity upon the Diffusion Coefficient of Radon Decay Products. *Health Phys.*, 37, 191.
5. Busigin, C., Busigin, A. and Phillips, C.R. (1982). The Chemical Fate of  $^{218}\text{Po}$  in Air. *Proc. Int. Conf. on Radiation Hazards in Mining*, Golden, CO (Gomez, M., ed.), Soc. Of Mining Engineers, Inc., 1043.
6. Goldstein, S.D. and Hopke, P.K. (1985). Environmental Neutralization of Polonium-218. *Environ. Sci. & Tech.*, 19, 146.
7. Chu, K.D. and Hopke, P.K. (1988). Neutralization Kinetics for Polonium-218. *Environ. Sci. & Technol.*, 22, 711.
8. Leung, H. M-Y. and Phillips, C.R. (1987). The Electrical and Diffusive Properties of Unattached  $^{218}\text{Po}$  in Argon Gas. *Radiation Protection Dosimetry*, 18(1), 3-11.
9. CRC Handbook of Chemistry and Physics, 66th ed., 85-86 Weast.
10. Luus, R. and Jaakola, T.H.I. (1973). Optimization by Direct Search and Systematic Reduction of the Size of Search Region. *A.I.Ch.E. J.*, 19, 760.
11. Fuchs, N.A. (1964). *The Mechanics of Aerosols*. Pergamon Press Limited, Oxford, England.
12. Nolan, P.J. (1943). *Proc. R. Ir. Acad., Sect. A*, 49, 67.

TABLE 1

Electrostatic and Diffusive Data:  $D_A$ , F, B and K for Po-218 Ions  
in the Propylene-Nitrogen Gas System

|     | $C_{Rn}$           | $C_{gas}$ | R.H. | F    | $D_A$ | K/B   | B    | K    |
|-----|--------------------|-----------|------|------|-------|-------|------|------|
| 1.  | $4.46 \times 10^6$ | 10        | 100  | 0.31 | 0.024 | 78.89 | 1.08 | 85.2 |
| 2.  | $1.15 \times 10^6$ | 10        | 97   | 0.41 | 0.019 | 56.28 | 1.21 | 68.1 |
| 3.  | $2.26 \times 10^5$ | 10        | 98   | 0.43 | 0.021 | 40.08 | 1.26 | 50.5 |
| 4.  | $1.08 \times 10^5$ | 10        | 100  | 0.57 | 0.025 | 32.28 | 1.23 | 39.7 |
| 5.  | $2.73 \times 10^4$ | 10        | 96   | 0.51 | 0.027 | 25.80 | 1.31 | 33.8 |
| 6.  | $5.89 \times 10^6$ | 10        | 45   | 0.61 | 0.042 | 56.11 | 1.49 | 83.6 |
| 7.  | $3.08 \times 10^6$ | 10        | 41   | 0.61 | 0.028 | 47.85 | 1.44 | 68.9 |
| 8.  | $5.21 \times 10^5$ | 10        | 40   | 0.67 | 0.038 | 32.51 | 1.52 | 49.4 |
| 9.  | $2.97 \times 10^5$ | 10        | 40   | 0.66 | 0.113 | 26.11 | 1.54 | 40.2 |
| 10. | $9.56 \times 10^4$ | 10        | 42   | 0.55 | 0.041 | 20.26 | 1.51 | 30.6 |
| 11. | $5.37 \times 10^4$ | 10        | 38   | 0.64 | 0.017 | 19.87 | 1.50 | 29.8 |
| 12. | $4.78 \times 10^6$ | 10        | 17   | 0.69 | 0.058 | 33.63 | 1.79 | 60.2 |
| 13. | $2.11 \times 10^5$ | 10        | 15   | 0.67 | 0.109 | 13.73 | 1.77 | 24.3 |
| 14. | $1.68 \times 10^5$ | 10        | 12   | 0.89 | 0.027 | 14.41 | 1.86 | 26.8 |
| 15. | $1.37 \times 10^5$ | 10        | 16   | 0.67 | 0.034 | 14.68 | 1.71 | 25.1 |
| 16. | $1.53 \times 10^4$ | 10        | 17   | 0.71 | 0.036 | 9.82  | 1.68 | 16.5 |

$C_{Rn}$  = radon concentration (atom/cm<sup>3</sup>)

$C_{gas}$  = propylene concentration (ppm)

R.H. = relative humidity (%)

F = fraction born charged

$D_A$  = diffusion coefficient (cm<sup>2</sup>/s)

K/B = neutralization rate constant to mobility ratio (V/cm<sup>2</sup>)

B = mobility (cm<sup>2</sup>/s.V)

K = neutralization rate constant (s<sup>-1</sup>)

TABLE 2

Diffusion Coefficient of the Po-218 Ion  
in the Propylene-Nitrogen Gas System

| Relative Humidity<br>R.H. (%) | Diffusivity<br>$D_A$ (cm <sup>2</sup> /s) |
|-------------------------------|-------------------------------------------|
| 100                           | 0.024                                     |
| 97                            | 0.019                                     |
| 98                            | 0.021                                     |
| 100                           | 0.025                                     |
| 96                            | 0.027                                     |
| 45                            | 0.042                                     |
| 41                            | 0.028                                     |
| 40                            | 0.038                                     |
| 40                            | 0.113                                     |
| 42                            | 0.041                                     |
| 38                            | 0.017                                     |
| 17                            | 0.058                                     |
| 15                            | 0.109                                     |
| 12                            | 0.027                                     |
| 16                            | 0.034                                     |
| 17                            | 0.036                                     |

Overall average = 0.041 cm<sup>2</sup>/s.

Average  $D_A$  at R.H. = 96-100% is 0.023 cm<sup>2</sup>/s.

TABLE 3

Fraction Born Charged of the Po-218 Ion  
in the Propylene-Nitrogen Gas System  
(Radon Concentration  $\sim 10^5$ , Fig. 1)

| Relative Humidity<br>R.H. (%) | Fraction Born Charged<br>f |
|-------------------------------|----------------------------|
| 98                            | 0.43                       |
| 42                            | 0.55                       |
| 16                            | 0.67                       |

TABLE 4

Mobility of the Po-218 Ion  
in the Propylene-Nitrogen Gas System  
(Radon Concentration  $\sim 10^5$ , Fig. 2)

| Relative Humidity<br>R.H. (%) | Mobility<br>B ( $\text{cm}^2/\text{s.V}$ ) |
|-------------------------------|--------------------------------------------|
| 100                           | 1.23                                       |
| 97                            | 1.21                                       |
| 42                            | 1.51                                       |
| 40                            | 1.54                                       |
| 16                            | 1.71                                       |

TABLE 5

Neutralization Rate Constant for Po-218 in the  
Propylene-Nitrogen Gas System  
(Propylene concentration = 10 ppm, Fig. 3)

| Relative Humidity<br>R.H. (%) | Radon Concentration<br>$C_{Rn}$ (atoms/cm <sup>3</sup> ) | Neutralization Rate Constant<br>$K$ (s <sup>-1</sup> ) |
|-------------------------------|----------------------------------------------------------|--------------------------------------------------------|
| 100                           | $4.46 \times 10^6$                                       | 85.2                                                   |
| 97                            | $1.15 \times 10^6$                                       | 68.1                                                   |
| 98                            | $2.26 \times 10^5$                                       | 50.5                                                   |
| 100                           | $1.08 \times 10^5$                                       | 39.7                                                   |
| 96                            | $2.73 \times 10^4$                                       | 33.8                                                   |
| 45                            | $5.89 \times 10^6$                                       | 83.6                                                   |
| 41                            | $3.08 \times 10^6$                                       | 68.9                                                   |
| 40                            | $5.21 \times 10^5$                                       | 49.4                                                   |
| 40                            | $2.97 \times 10^5$                                       | 40.2                                                   |
| 42                            | $9.56 \times 10^4$                                       | 30.6                                                   |
| 38                            | $5.37 \times 10^4$                                       | 29.8                                                   |
| 17                            | $4.78 \times 10^6$                                       | 60.2                                                   |
| 15                            | $2.11 \times 10^5$                                       | 24.3                                                   |
| 12                            | $1.68 \times 10^5$                                       | 26.8                                                   |
| 16                            | $1.37 \times 10^5$                                       | 25.1                                                   |
| 17                            | $1.53 \times 10^4$                                       | 16.5                                                   |

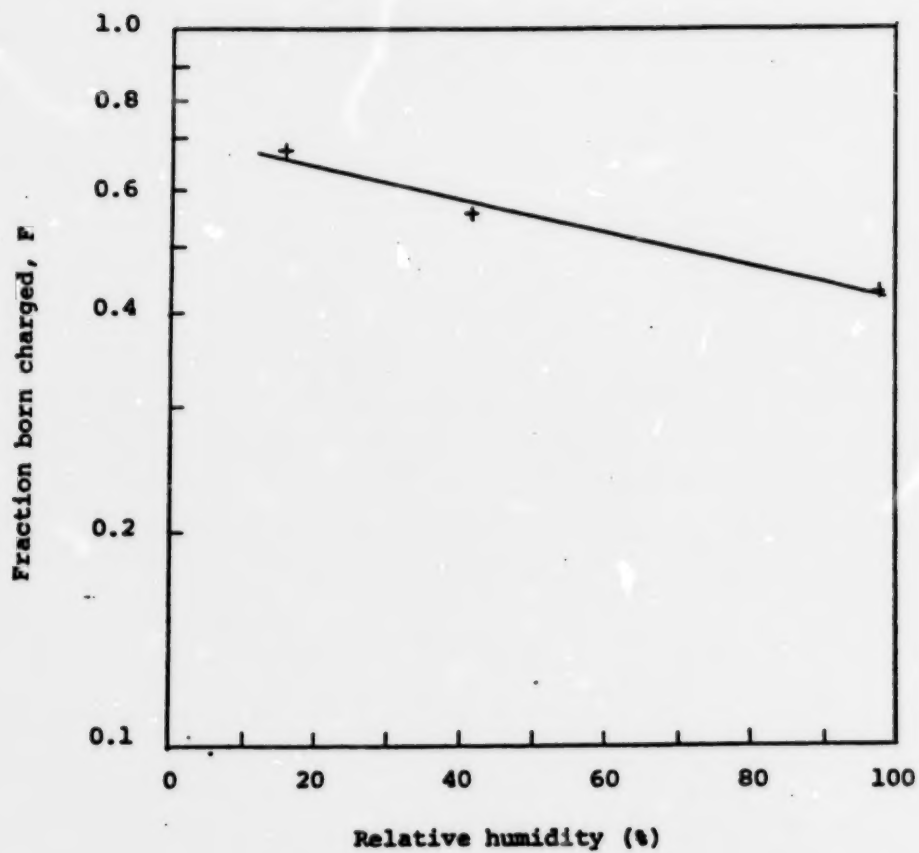


Fig. 1. Effect of relative humidity on fraction born charged of Po-218, in a 10 ppm propylene (balance nitrogen) system.

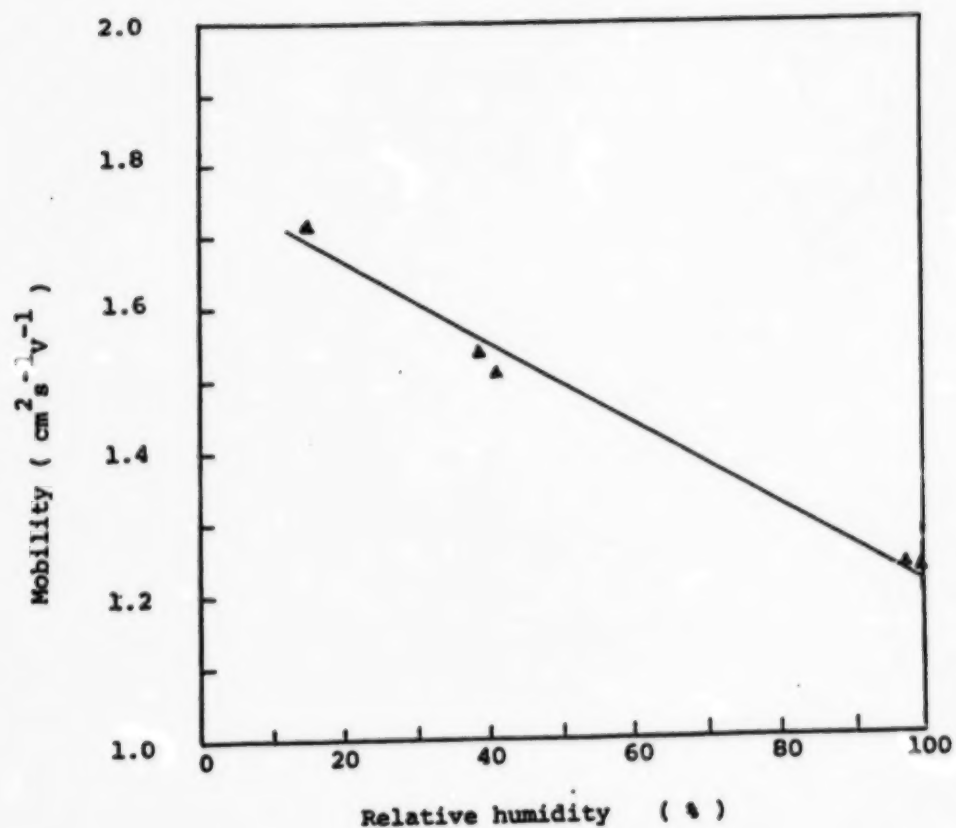


Fig. 2. Effect of relative humidity on the mobility of Po-218, in a 10 ppm propylene (balance nitrogen) system.

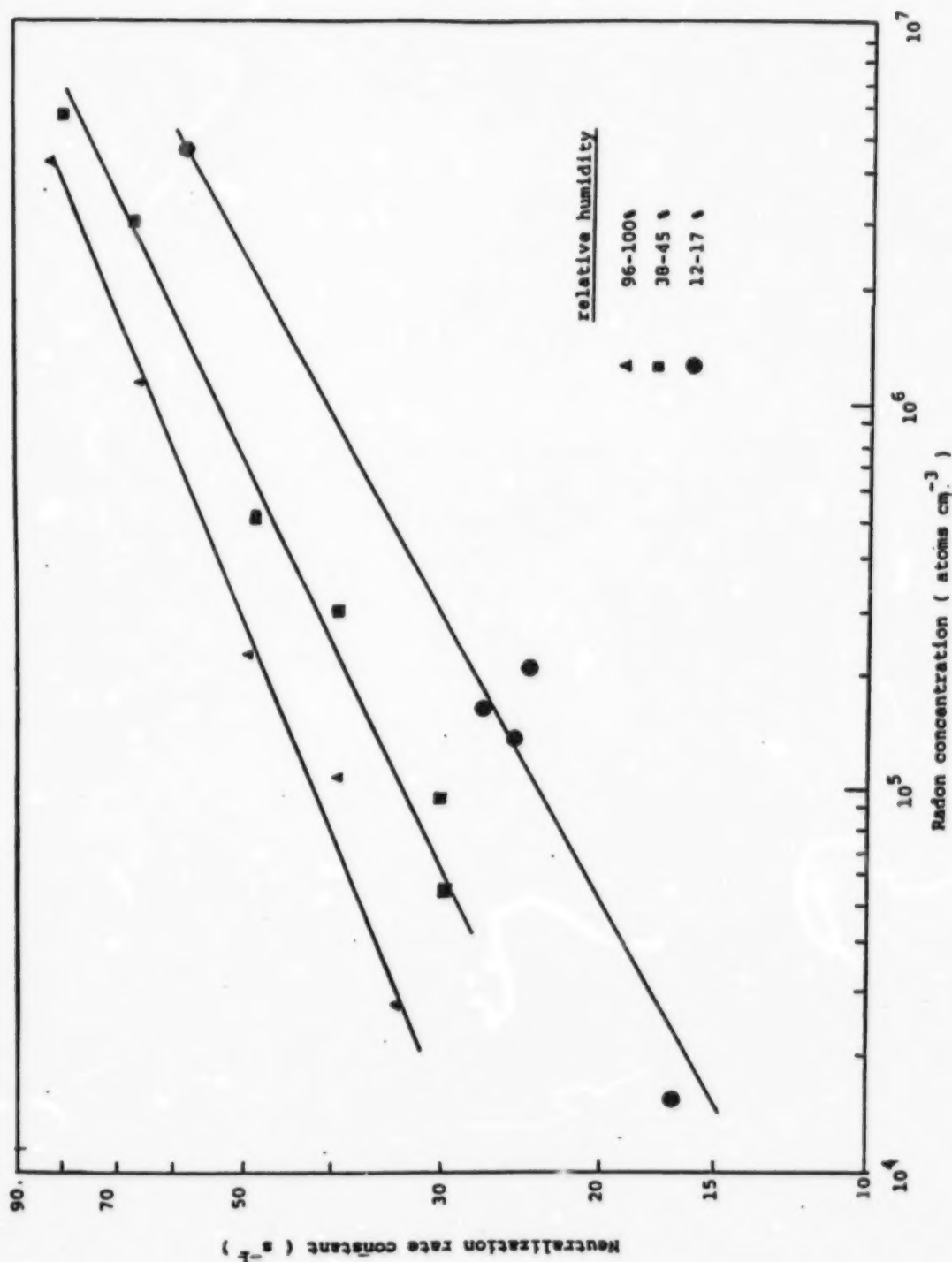


Fig. 3. Effect of radon concentration and relative humidity on the neutralization rate constant of Po-218, in a 10 ppm propylene (balance nitrogen) system.